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(19) (CA) **APPLICATION FOR CANADIAN PATENT** (12)

(54) Diarylpyrazolinones

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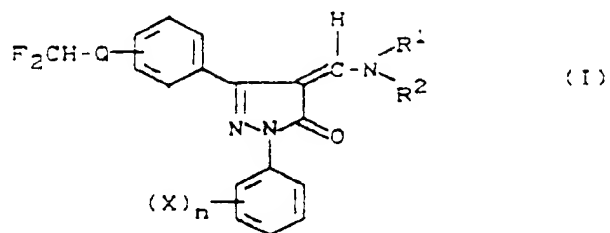
Notice: The specification contained herein as filed

**Canada**

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## Abstract

The invention relates to new diarylpyrazolinones of the general formula (I)

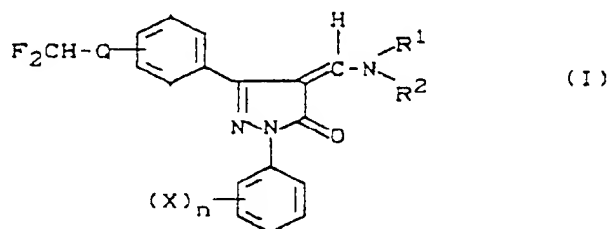


in which  $n$ ,  $Q$ ,  $X$ ,  $R^1$  and  $R^2$  have the meanings given in the description, to two processes for their preparation, to various new intermediates, and to their use as herbicides.

The invention relates to new diarylpyrazolinones, to two processes for their preparation, to various new intermediates, and to their use as herbicides.

5 It has already been disclosed that certain pyrazolin-5-one derivatives such as, for example, 5-(3-methoxyphenyl)-4-methylaminomethylene-2-phenyl-2,4-dihydro-3H-pyrazol-3-one have herbicidal properties (cf. EP-A 274,642). However, the herbicidal action of the pyrazolin-5-one derivatives which are known to date is not  
10 always entirely satisfactory.

There have now been found new diarylpyrazolinones of the general formula (I)



in which

15 n represents the numbers 0, 1, 2 or 3,

Q represents oxygen or sulphur,

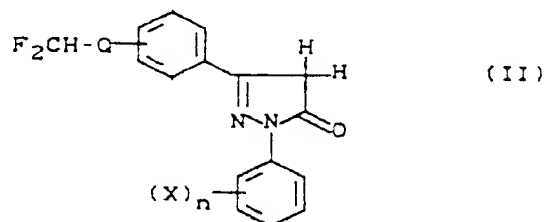
- R<sup>1</sup> represents hydrogen or C<sub>1</sub>-C<sub>4</sub>-alkyl,
- R<sup>2</sup> represents hydrogen, hydroxyl, amino, or a radical from the series comprising C<sub>1</sub>-C<sub>6</sub>-alkyl, C<sub>2</sub>-C<sub>6</sub>-alkenyl, C<sub>2</sub>-C<sub>6</sub>-alkinyl, C<sub>1</sub>-C<sub>6</sub>-hydroxyalkyl, C<sub>1</sub>-C<sub>4</sub>-alkoxy-C<sub>1</sub>-C<sub>4</sub>-alkyl, C<sub>3</sub>-C<sub>6</sub>-cycloalkyl-C<sub>1</sub>-C<sub>4</sub>-alkyl, phenyl-C<sub>1</sub>-C<sub>4</sub>-alkyl, C<sub>1</sub>-C<sub>6</sub>-alkoxy, C<sub>3</sub>-C<sub>6</sub>-alkenyloxy, phenyl-C<sub>1</sub>-C<sub>2</sub>-alkoxy, C<sub>1</sub>-C<sub>6</sub>-alkylamino, C<sub>1</sub>-C<sub>4</sub>-alkylcarbonylamino or di-(C<sub>1</sub>-C<sub>4</sub>-alkyl)-amino, each of which is optionally substituted by halogen, and
- 5 X represents hydrogen, halogen, or a radical from the series comprising C<sub>1</sub>-C<sub>4</sub>-alkyl or C<sub>1</sub>-C<sub>4</sub>-alkoxy, each of which is optionally substituted by halogen, the following compounds - disclosed in DE-OS (German Published Specification) 3,941,240, p. 44, 45, 95 and 96 - being excepted by disclaimer:
- 10 2-phenyl-, 2-(4-fluorophenyl)- and 2-(4-chlorophenyl)-5-(2-difluoromethylthiophenyl)-4-(N-hydroxymethylaminomethylene)-2,4-dihydro-3H-pyrazol-3-one, 2-phenyl-, 2-(4-fluorophenyl)- and 2-(4-chlorophenyl)-5-(3-difluoromethylthiophenyl)-4-(N-hydroxymethylaminomethylene)-2,4-dihydro-3H-pyrazol-3-one, 2-phenyl-, 2-(4-fluorophenyl)- and 2-(4-chlorophenyl)-5-(2-difluoromethylthiophenyl)-4-dimethylaminomethylene-2,4-dihydro-3H-pyrazol-3-one as well as 2-phenyl-, 2-(4-fluorophenyl)- and 2-(4-chlorophenyl)-5-(3-difluoromethylthiophenyl)-4-dimethylaminomethylene-2,4-dihydro-3H-pyrazol-3-one.
- 20
- 25

If appropriate, the compounds of the formula (I) can exist in various stereoisomeric or tautomeric forms. The invention relates to the pure isomers as well as to the mixtures of these isomers. For simplicity's sake, the following text will always refer to compounds of the formula (I), this being understood as meaning the pure isomers as well as the various mixtures of these isomers which are possible.

The new compounds of the general formula (I) are obtained when

- (a) in the event that, in formula (I),  $R^1$  and  $R^2$  represent methyl and n, Q and X have the abovementioned meanings,

pyrazolinones of the general formula (II)



in which

n, Q and X have the abovementioned meanings,

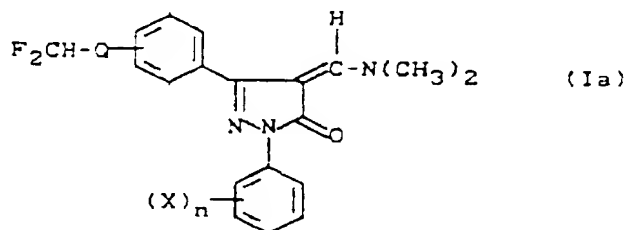
are reacted with dimethylformamide acetals of the general formula (III)



in which

5        R        represents  $\text{C}_1\text{-C}_4\text{-alkyl}$  or benzyl,  
if appropriate in the presence of a diluent, or when

(b) diarylpyrazolinones of the general formula (Ia)



in which

10        n, Q and X have the abovementioned meanings,  
are reacted with amines of the general formula (IV)



in which

$R^1$  and  $R^2$  have the abovementioned meanings,

if appropriate in the presence of a diluent.

5 The diarylpyrazolinones of the general formula (I) according to the invention are distinguished by a powerful herbicidal activity.

10 Surprisingly, the new compounds of the general formula (I) show a considerably better herbicidal action than the previously known pyrazolin-5-one derivatives, which are comparable substances from the point of view of their structure and profile of action.

The invention preferably relates to compounds of the formula (I) in which

n represents the numbers 0, 1 or 2,

15 Q represents oxygen or sulphur,

$R^1$  represents hydrogen, methyl or ethyl,

20  $R^2$  represents hydrogen, hydroxyl, amino, or represents a radical from the series comprising  $C_1$ - $C_5$ -alkyl,  $C_3$ - $C_5$ -alkenyl,  $C_3$ - $C_5$ -alkinyl,  $C_1$ - $C_5$ -hydroxyalkyl,  $C_1$ - $C_2$ -alkoxy- $C_1$ - $C_2$ -alkyl,  $C_1$ - $C_4$ -alkoxy or  $C_3$ - $C_4$ -alkenyloxy, each of which is optionally substituted

by fluorine and/or chlorine;

5 or represents a radical from the series comprising  $C_1$ - $C_6$ -cycloalkyl- $C_1$ - $C_2$ -alkyl, phenyl- $C_1$ - $C_2$ -alkyl, phenyl- $C_1$ - $C_2$ -alkoxy,  $C_1$ - $C_6$ -alkylamino,  $C_1$ - $C_6$ -alkyl-carbonylamino or dimethylamino, each of which is optionally substituted by fluorine, chlorine and/or bromine, and

10 X represents hydrogen, fluorine, chlorine, bromine or a radical from the series comprising methyl, ethyl, methoxy or ethoxy, each of which is optionally substituted by fluorine and/or chlorine, with the exception of the compounds being excepted by disclaimer.

15 In particular, the invention relates to compounds of the formula (I) in which

n represents the numbers 0, 1 or 2,

Q represents oxygen or sulphur,

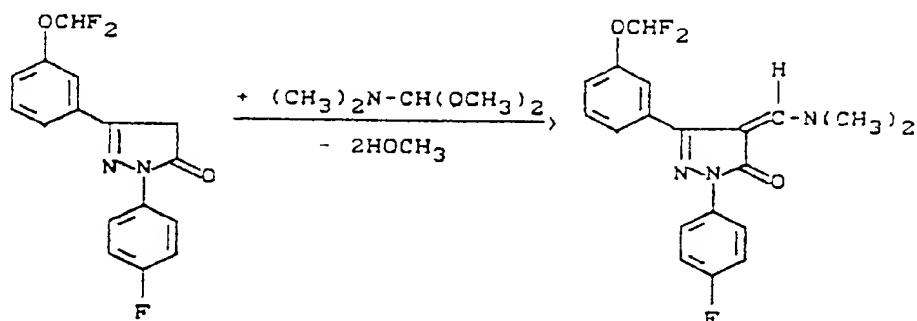
$R^1$  represents hydrogen or methyl,

20  $R^2$  represents hydrogen, hydroxyl, amino, or represents a radical from the series comprising methyl, ethyl, propyl, isopropyl, butyl, isobutyl, sec-butyl or tert-butyl, each of which is optionally substituted by fluorine, or represents allyl, propargyl,

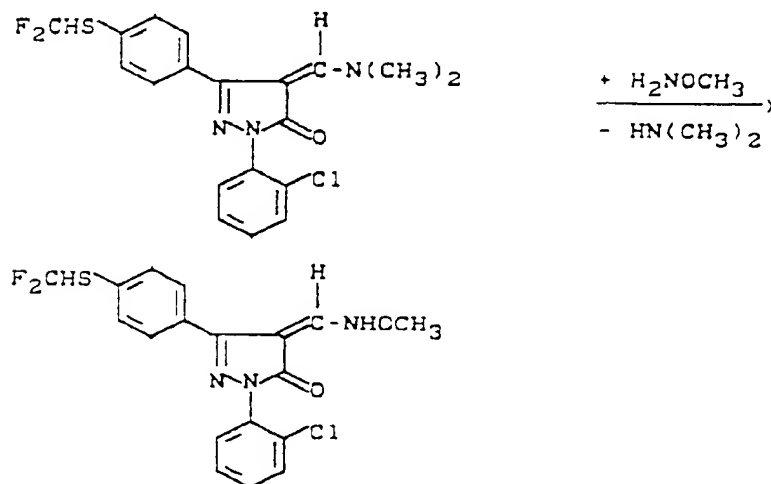
1-methyl-propargyl, 1,1-dimethylpropargyl, hydroxy-ethyl, hydroxypropyl, methoxyethyl, ethoxyethyl, cyclopropylmethyl, cyclohexylmethyl, cyclohexylethyl, phenylmethyl, phenylethyl, methoxy, ethoxy, propoxy, allyloxy, benzyloxy, methylamino, dimethylamino, acetylamino, propionylamino, butyrylamino or isobutyrylamino, and

X represents hydrogen, fluorine, chlorine or trifluoromethyl, with the exception of the compounds being excepted by disclaimer.

If, for example, 1-(4-fluoro-phenyl)-3-(3-difluoromethoxyphenyl)-pyrazolin-5-one and dimethylformamide dimethyl acetal are used as starting substances, the course of the reaction in process (a) according to the invention can be outlined by the following equation:



If, for example, 1-(2-chlorophenyl)-3-(4-difluoromethylthiophenyl)-4-dimethylaminomethylene-pyrazolin-5-one and O-methyl-hydroxylamine are used as starting substances, the course of the reaction in process (b) according to the invention can be outlined by the following equation:

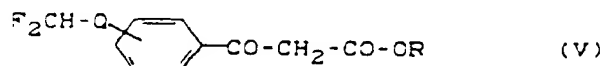


Formula (II) provides a general definition of the pyrazolinones to be used as starting substances in process (a) according to the invention for the preparation of compounds of the formula (I).

In formula (II), n, Q and X preferably, or in particular, have those meanings which have already been mentioned above in connection with the description of the compounds of the formula (I) according to the invention as being preferred, or particularly preferred, for n, Q and X.

The pyrazolinones of the formula (II) were hitherto unknown from the literature and, being new substances, are also a subject of the present patent application.

5 The new pyrazolinones of the formula (II) are obtained when aroylacetaes of the general formula (V)

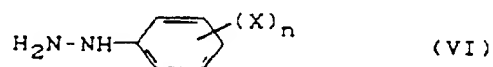


in which

Q represents oxygen or sulphur and

R represents C<sub>1</sub>-C<sub>6</sub>-alkyl, preferably methyl or ethyl,

10 are reacted with arylhydrazines of the general formula (VI)



in which

n represents the numbers 0, 1, 2 or 3 and

15 X represents hydrogen, halogen, or a radical from the series comprising C<sub>1</sub>-C<sub>4</sub>-alkyl or C<sub>1</sub>-C<sub>4</sub>-alkoxy, each of which is optionally substituted by halogen,

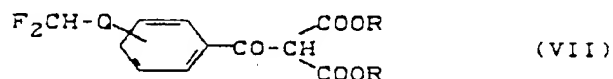
at temperatures between -20°C and +80°C, if appropriate

in the presence of a diluent such as, for example, ethanol, and if appropriate in the presence of a reaction auxiliary such as, for example, sodium acetate, and the product is worked up by customary methods (cf. the Preparation Examples).

The arylhydrazines of the formula (VI) are known and/or can be prepared by processes known per se (cf. US Patent 4,411,839; DE-OS (German Published Specification) 1,927,924; Khim. Farm. Zh. 10 (1976), 27-31 - cited in Chem. Abstracts 86: 139926a).

The aroylacetaes of the formula (V) were hitherto unknown from the literature and, being new substances, are also a subject of the present patent application.

The new aroylacetaes of the formula (V) are obtained when corresponding aroylmalonic diesters of the general formula (VII)



in which

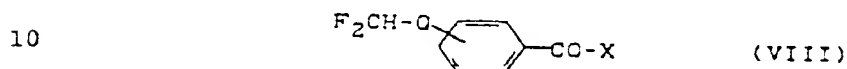
Q represents oxygen or sulphur and

R represents C<sub>1</sub>-C<sub>6</sub>-alkyl, preferably methyl or ethyl, are refluxed with water in the presence of a catalyst

such as, for example, p-toluenesulphonic acid, and the product is then worked up by customary methods (cf. the Preparation Examples).

5 The aroylmalonic diesters of the formula (VII) were hitherto unknown from the literature and, being new substances, are also a subject of the present patent application.

The new aroylmalonic diesters of the formula (VII) are obtained when aroyl halides of the general formula (VIII)

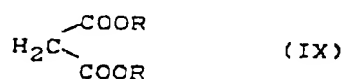


in which

Q represents oxygen or sulphur and

X represents halogen, in particular fluorine, chlorine or bromine,

15 are reacted with malonic diesters of the general formula (IX)



in which

R represents C<sub>1</sub>-C<sub>8</sub>-alkyl, preferably methyl or ethyl,

at temperatures between -20°C and +50°C, in the presence of a diluent such as, for example, acetonitrile, in the presence of a reaction auxiliary such as, for example, magnesium chloride, and in the presence of an acid binder such as, for example, triethylamine, and the product is worked up by customary methods (cf. the Preparation Examples).

The aroyl halides of the formula (VIII) are known and/or can be prepared by processes known per se (cf. US Patent Specification 4,832,879; JP 59,181,259 - cited in Chem. Abstracts 102: 113480z; DE-OS (German Published Specification) 2,914,915; US Patent Specification 4,009,208; US Patent Specification 3,960,945; US Patent Specification 3,895,036; Ukr. Khim. Zh. 47 (1981), 871-874 - cited in Chem. Abstracts 95: 186790x).

The malonic diesters of the formula (IX) are known chemicals for synthesis.

Formula (Ia) provides a general definition of the diaryl-pyrazolinones to be used as starting substances in process (b) according to the invention for the preparation of compounds of the formula (I).

In formula (Ia), n, Q and X preferably, or in particular, have those meanings which have already been mentioned above in connection with the description of the compounds

of the formula (I) according to the invention as being preferred, or particularly preferred, for n, Q and X.

5 The starting substances of the formula (Ia) are a subset of the diarylpyrazolinones of the formula (I) according to the invention; they can be prepared by process (a) according to the invention.

Formula (IV) provides a general definition of the amines furthermore to be used as starting substances in process (b) according to the invention.

10 In formula (IV),  $R^1$  and  $R^2$  preferably, or in particular, have those meanings which have already been mentioned above in connection with the description of the compounds of the formula (I) according to the invention as being preferred, or particularly preferred, for  $R^1$  and  $R^2$ .

15 The starting substances of the formula (IV) are known chemicals for synthesis.

Processes (a) and (b) according to the invention for the preparation of the new diarylpyrazolinones of the formula (I) are preferably carried out using diluents. Diluents which are suitable for this purpose are all inert organic solvents. These preferably include aliphatic and aromatic, optionally halogenated hydrocarbons such as pentane, hexane, heptane, cyclohexane, petroleum ether, benzine, ligroin, benzene, toluene, xylene, methylene chloride, ethylene chloride, chloroform, carbon

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tetrachloride, chlorobenzene and o-dichlorobenzene, ethers such as diethyl ether and dibutyl ether, glycol dimethyl ether and diglycol dimethyl ether, tetrahydrofuran and dioxane, alcohols such as methanol, ethanol and isopropanol, ketones such as acetone, methyl ethyl ketone, methyl isopropyl ketone and methyl isobutyl ketone, esters such as methyl acetate and ethyl acetate, nitriles such as, for example, acetonitrile and propionitrile, amides such as, for example, dimethylformamide, dimethylacetamide and N-methyl-pyrrolidone, and also dimethyl sulphoxide, tetramethylene sulphone and hexamethylphosphoric triamide.

When carrying out process (a) and (b) according to the invention, the reaction temperatures can be varied within a substantial range. In general, the process is carried out at temperatures between 0°C and 100°C, preferably at temperatures between 10°C and 50°C.

Processes (a) and (b) according to the invention are generally carried out under atmospheric pressure. However, it is also possible to carry out the process under increased or reduced pressure.

For carrying out processes (a) and (b) according to the invention, the starting substances required in each case are generally employed in approximately equimolar amounts. However, it is also possible to use one of the two components employed in each case in a larger excess. In general, the reactions are carried out in a suitable

diluent, and the reaction mixture is stirred for several hours at the temperature required in each case. Working-up is carried out in each case by customary methods (cf. the Preparation Examples).

- 5      The active compounds according to the invention can be used as defoliants, desiccants, agents for destroying broad-leaved plants and, especially, as weed-killers. By weeds, in the broadest sense, there are to be understood all plants which grow in locations where they are  
10      undesired. Whether the substances according to the invention act as total or selective herbicides depends essentially on the amount used.

15      The active compounds according to the invention can be used, for example, in connection with the following plants:

20      Dicotyledon weeds of the genera: Sinapis, Lepidium, Galium, Stellaria, Matricaria, Anthemis, Galinsoga, Chenopodium, Urtica, Senecio, Amaranthus, Portulaca, Xanthium, Convolvulus, Ipomoea, Polygonum, Sesbania, Ambrosia, Cirsium, Carduus, Sonchus, Solanum, Rorippa, Rotala, Lindernia, Lamium, Veronica, Abutilon, Emex, Datura, Viola, Galeopsis, Papaver, Centaurea, Trifolium, Ranunculus and Taraxacum.

25      Dicotyledon cultures of the genera: Gossypium, Glycine, Beta, Daucus, Phaseolus, Pisum, Solanum, Linum, Ipomoea, Vicia, Nicotiana, Lycopersicon, Arachis, Brassica,

Lactuca, Cucumis and Cucurbita.

Monocotyledon weeds of the genera: Echinochloa, Setaria, Panicum, Digitaria, Phleum, Poa, Festuca, Eleusine, Brachiaria, Lolium, Bromus, Avena, Cyperus, Sorghum, Agropyron, Cynodon, Monochoria, Fimbristylis, Sagittaria, Eleocharis, Scirpus, Paspalum, Ischaemum, Sphenoclea, Dactyloctenium, Agrostis, Alopecurus and Apera.

Monocotyledon cultures of the genera: Oryza, Zea, Triticum, Hordeum, Avena, Secale, Sorghum, Panicum, Saccharum, Ananas, Asparagus and Allium.

However, the use of the active compounds according to the invention is in no way restricted to these genera, but also extends in the same manner to other plants.

The compounds are suitable, depending on the concentration, for the total combating of weeds, for example on industrial terrain and rail tracks, and on paths and squares with or without tree plantings. Equally, the compounds can be employed for combating weeds in perennial cultures, for example afforestations, decorative tree plantings, orchards, vineyards, citrus groves, nut orchards, banana plantations, coffee plantations, tea plantations, rubber plantations, oil palm plantations, cocoa plantations, soft fruit plantings and hopfields, in lawns, turf and pasture-land, and for the selective combating of weeds in annual cultures.

5 The compounds of the formula (I) according to the invention are particularly suitable for selectively combating monocotyledon and dicotyledon weeds in monocotyledon and dicotyledon cultures, both by the pre- and the post-emergence method.

10 The active compounds can be converted into the customary formulations, such as solutions, emulsions, wettable powders, suspensions, powders, dusting agents, pastes, soluble powders, granules, suspension-emulsion concentrates, natural and synthetic materials impregnated with active compound, and very fine capsules in polymeric substances.

15 These formulations are produced in a known manner, for example by mixing the active compounds with extenders, that is liquid solvents and/or solid carriers, optionally with the use of surface-active agents, that is emulsifying agents and/or dispersing agents and/or foam-forming agents.

20 In the case of the use of water as an extender, organic solvents can, for example, also be used as auxiliary solvents. As liquid solvents, there are suitable in the main: aromatics, such as xylene, toluene, or alkyl-naphthalenes, chlorinated aromatics and chlorinated aliphatic hydrocarbons, such as chlorobenzenes, chloroethylenes or  
25 methylene chloride, aliphatic hydrocarbons, such as cyclohexane or paraffins, for example petroleum fractions, mineral and vegetable oils, alcohols, such as

butanol or glycol as well as their ethers and esters, ketones, such as acetone, methyl ethyl ketone, methyl isobutyl ketone or cyclohexanone, strongly polar solvents, such as dimethylformamide and dimethyl sulphoxide, as well as water.

5

As solid carriers there are suitable: for example ammonium salts and ground natural minerals, such as kaolins, clays, talc, chalk, quartz, attapulgit, montmorillonite or diatomaceous earth, and ground synthetic minerals, such as highly disperse silica, alumina and silicates; as solid carriers for granules there are suitable: for example crushed and fractionated natural rocks such as calcite, marble, pumice, sepiolite and dolomite, as well as synthetic granules of inorganic and organic meals, and granules of organic material such as sawdust, coconut shells, maize cobs and tobacco stalks; as emulsifying and/or foam-forming agents there are suitable: for example non-ionic and anionic emulsifiers, such as polyoxyethylene fatty acid esters, polyoxyethylene fatty alcohol ethers, for example alkylaryl polyglycol ethers, alkylsulphonates, alkyl sulphates, arylsulphonates as well as albumen hydrolysis products; as dispersing agents there are suitable: for example lignin-sulphite waste liquors and methylcellulose.

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Adhesives such as carboxymethylcellulose and natural and synthetic polymers in the form of powders, granules or latexes, such as gum arabic, polyvinyl alcohol and polyvinyl acetate, as well as natural phospholipids, such

as cephalins and lecithins, and synthetic phospholipids, can be used in the formulations. Further additives can be mineral and vegetable oils.

- 5 It is possible to use colorants such as inorganic pigments, for example iron oxide, titanium oxide and Prussian Blue, and organic dyestuffs, such as alizarin dyestuffs, azo dyestuffs and metal phthalocyanine dyestuffs, and trace nutrients such as salts of iron, manganese, boron, copper, cobalt, molybdenum and zinc.
- 10 The formulations in general contain between 0.1 and 95 per cent by weight of active compound, preferably between 0.5 and 90%.
- 15 For combating weeds, the active compounds according to the invention, as such or in the form of their formulations, can also be used as mixtures with known herbicides, finished formulations or tank mixes being possible.
- 20 Suitable herbicides for the mixtures are known herbicides such as anilides, such as, for example, diflufenican and propanil; arylcarboxylic acids such as, for example, dichloropicolinic acid, dicamba and picloram; aryloxy-alkanoic acids such as, for example, 2,4 D, 2,4 DB, 2,4 DP, fluroxypyr, MCPA, MCPP and triclopyr; aryloxy-phenoxy-alkanoates such as, for example, diclofop-methyl,
- 25 fenoxaprop-ethyl, fluazifop-butyl, haloxyfop-methyl and quizalofop-ethyl; azinones such as, for example,

chloridazon and norflurazon; carbamates such as, for  
 example, chlorpropham, desmedipham, phenmedipham and  
 propham; chloroacetanilides such as, for example,  
 alachlor, acetochlor, butachlor, metazachlor,  
 5 metolachlor, pretilachlor and propachlor; dinitroanilines  
 such as, for example, oryzalin, pendimethalin and tri-  
 fluralin; diphenyl ethers such as, for example,  
 acifluorfen, bifenox, fluoroglycofen, fomesafen, halo-  
 safen, lactofen and oxyfluorfen; ureas such as, for  
 10 example, chlortoluron, diuron, fluometuron, isoproturon,  
 linuron and methabenzthiazuron; hydroxylamines such as,  
 for example, alloxydim, clethodim, cycloxydim, sethoxydim  
 and tralkoxydim; imidazolinones such as, for example,  
 imazethapyr, imazamethabenz, imazapyr and imazaquin;  
 15 nitriles such as, for example, bromoxynil, dichlobenil  
 and ioxynil; oxyacetamides such as, for example,  
 mefenacet; sulphonylureas such as, for example, amido-  
 sulfuron, bensulfuron-methyl, chlorimuron-ethyl, chlor-  
 sulfuron, cinosulfuron, metsulfuron-methyl, nicosulfuron,  
 20 primisulfuron, pyrazosulfuron-ethyl, thifensulfuron-  
 methyl, triasulfuron and tribenuron-methyl; thiocar-  
 bamates such as, for example, butylate, cycloate, di-  
 allate, EPTC, esprocarb, molinate, prosulfocarb, thio-  
 bencarb and tri-allate; triazines such as, for example,  
 25 atrazin, cyanazin, simazin, simetryne, terbutryne and  
 terbutylazin; triazinones such as, for example, hexa-  
 zinone, met amitron and metribuzin; others such as, for  
 example, aminotriazol, benfuresate, bentazone, cin-  
 methylin, clomazone, clopyralid, difenzoquat, dithiopyr,  
 30 ethofumesate, fluorchloridone, glufosinate, glyphosate,

isoxaben, pyridate, quinchlorac, quinmerac, sulphosate and tridiphane.

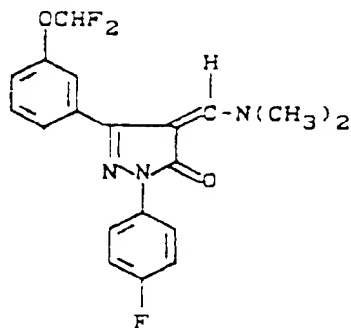
5 Mixtures with other known active compounds, such as fungicides, insecticides, acaricides, nematocides, bird repellants, plant nutrients and agents which improve soil structure, are also possible.

10 The active compounds can be used as such, in the form of their formulations or in the use forms prepared therefrom by further dilution, such as ready-to-use solutions, suspensions, emulsions, powders, pastes and granules. They are used in the customary manner, for example by watering, spraying, atomizing or scattering.

15 The active compounds according to the invention can be applied either before or after emergence of the plants. They can also be incorporated into the soil before sowing.

20 The amount of active compound used can vary within a substantial range. It depends essentially on the nature of the desired effect. In general, the amounts applied are between 1 g and 10 kg of active compound per hectare of soil surface, preferably between 5 g and 5 kg per ha.

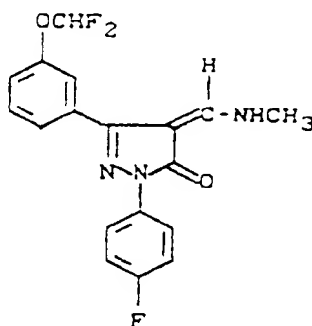
The preparation and use of the active compounds according to the invention can be seen from the following examples.

Preparation Examples:Example 1

(Process (a))

5 A mixture of 6.5 g (20 mmol) of 2-(4-fluorophenyl)-5-(3-difluoromethoxyphenyl)-4-dimethylaminomethylene-2,4-dihydro-3H-pyrazol-3-one, 2.7 g (22mmol) of dimethylformamide dimethyl acetal and 150 ml of toluene is stirred for 2 hours at 20°C. It is  
 10 then concentrated under a water pump vacuum, the residue is triturated with petroleum ether, and the product which is obtained as crystals is isolated by filtration.

6.9 g (92% of theory) of 2-(4-fluorophenyl)-5-(3-difluoromethoxyphenyl)-4-dimethylaminomethylene-2,4-dihydro-3H-pyrazol-3-one of melting point 103°C are  
 15 obtained.

Example 2

(Process (b))

5 A mixture of 2.8 g (7.5 mmol) of 2-(4-fluorophenyl)-5-(3-difluoromethoxyphenyl)-4-methylaminomethylene-2,4-dihydro-3H-pyrazol-3-one, 50 ml of methanol and 3 ml of a 30% aqueous methylamine solution (10 mmol  $\text{H}_2\text{NCH}_3$ ), is stirred for 3 hours at 20°C. It is then concentrated under a water pump vacuum, the residue is triturated with  
 10 petroleum ether, and the product which has been obtained as crystals is isolated by filtration.

2.4 g (88.5% of theory) of 2-(4-fluorophenyl)-5-(3-difluoromethoxyphenyl)-4-methylaminomethylene-2,4-dihydro-3H-pyrazol-3-one of melting point 187°C are  
 15 obtained.

Other examples of compounds of the formula (I) which can be prepared analogously to Preparation Examples 1 and 2 and following the general description of the preparation processes according to the invention are those listed in

Table 1 below.

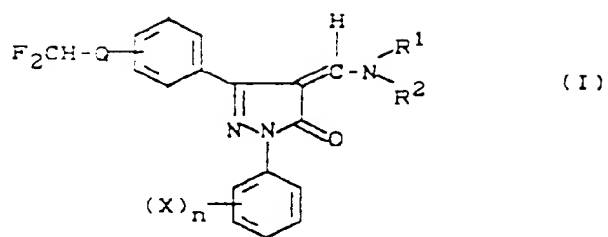


Table 1: Examples of the compounds of the formula (I)

Ex. No.	n	(Position) Q	R <sup>1</sup>	R <sup>2</sup>	(Position) X	Melting point (°C)
3	1	(2-)S	H	CH <sub>3</sub>	(4-)F	
4	2	(2-)S	CH <sub>3</sub>	CH <sub>3</sub>	(2,4-)F <sub>2</sub>	122
5	0	(3-)O	CH <sub>3</sub>	OH	-	111
6	1	(2-)S	CH <sub>3</sub>	CH <sub>3</sub>	(4-)CF <sub>3</sub>	93
7	1	(2-)S	H	CH <sub>3</sub>	(4-)CF <sub>3</sub>	139
8	1	(2-)S	H	NHCH <sub>3</sub>	(4-)F	125
9	1	(2-)S	H	N(CH <sub>3</sub> ) <sub>2</sub>	(4-)F	108
10	1	(2-)S	CH <sub>3</sub>	CH <sub>3</sub>	(3-)Cl	104
11	0	(2-)S	H	CH <sub>3</sub>	-	116
12	1	(2-)S	H	NHCH <sub>3</sub>	(4-)CF <sub>3</sub>	137
13	1	(2-)S	H	N(CH <sub>3</sub> ) <sub>2</sub>	(4-)CF <sub>3</sub>	107
14	1	(2-)S	CH <sub>3</sub>	OH	(4-)CF <sub>3</sub>	118
15	2	(2-)S	H	CH <sub>3</sub>	(2,4-)F <sub>2</sub>	75
16	0	(2-)S	H	NHCH <sub>3</sub>	-	
17	0	(2-)S	H	N(CH <sub>3</sub> ) <sub>2</sub>	-	
18	1	(3-)O	H	CH <sub>3</sub>	(2-)F	126
19	1	(2-)S	H	CH <sub>3</sub>	(3-)Cl	138
20	1	(2-)S	H	CH <sub>3</sub>	(4-)Cl	145
21	1	(3-)O	CH <sub>3</sub>	OH	(2-)F	78
22	2	(2-)S	CH <sub>3</sub>	OH	(2,4-)F <sub>2</sub>	141

Table 1 - Continuation

5

Ex. No.	n	(Position) Q	R <sup>1</sup>	R <sup>2</sup>	(Position) X	Melting point (°C)
23	1	(2-)S	H	CH <sub>3</sub>	(2-)F	127
24	1	(2-)S	H	NHCH <sub>3</sub>	(3-)Cl	88
25	0	(3-)O	H	CH <sub>3</sub>	-	137
26	1	(2-)S	CH <sub>3</sub>	OH	(3-)Cl	147
27	0	(3-)S	H	CH <sub>3</sub>	-	79
28	1	(3-)S	CH <sub>3</sub>	CH <sub>3</sub>	(3-)Cl	132
29	1	(3-)S	CH <sub>3</sub>	CH <sub>3</sub>	(4-)CF <sub>3</sub>	121
30	1	(3-)S	CH <sub>3</sub>	CH <sub>3</sub>	(3-)F	114
31	1	(3-)S	H	N(CH <sub>3</sub> ) <sub>2</sub>	(4-)F	
32	1	(3-)S	H	CH <sub>3</sub>	(4-)F	105
33	1	(2-)S	CH <sub>3</sub>	OH	(2-)F	
34	1	(3-)O	CH <sub>3</sub>	OH	(4-)F	
35	1	(3-)S	CH <sub>3</sub>	OH	(3-)Cl	105
36	1	(3-)S	CH <sub>3</sub>	OH	(3-)F	58
37	1	(3-)S	H	CH <sub>3</sub>	(3-)Cl	97
38	1	(3-)S	H	CH <sub>3</sub>	(3-)F	92
39	1	(2-)S	H	$\begin{array}{c} \text{CH}_3 \\   \\ -\text{C}-\text{C}\equiv\text{CH} \\   \\ \text{CH}_3 \end{array}$	(4-)F	99
40	1	(3-)S	H	CH <sub>3</sub>	(4-)CF <sub>3</sub>	127

Table 1 - Continuation

5

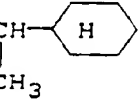

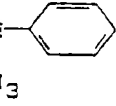


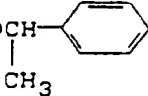
Ex. No.	n	(Position) Q	R <sup>1</sup>	R <sup>2</sup>	(Position) X	Melting point (°C)
41	1	(3-)S	CH <sub>3</sub>	OH	(4-)CF <sub>3</sub>	135
42	1	(3-)S	H	NHCH <sub>3</sub>	(3-)Cl	
43	1	(2-)S	H	(R)- 	(4-)F	49
44	1	(2-)S	H	-CH <sub>2</sub> C(CH <sub>3</sub> ) <sub>3</sub>	(4-)F	105
45	1	(2-)S	H	-CH <sub>2</sub> 	(4-)F	84
46	1	(2-)S	H	-CH <sub>2</sub> -C≡CH	(4-)F	158
47	1	(2-)S	H	C <sub>4</sub> H <sub>9</sub>	(4-)F	
48	1	(2-)S	H	(R)- 	(4-)F	58
49	1	(2-)S	H	-CH <sub>2</sub> -CH 	(4-)F	127
50	1	(2-)S	H	OCH <sub>3</sub>	(4-)F	46
51	1	(2-)S	H	-OCH <sub>2</sub> 	(4-)F	
52	1	(2-)S	H	-CH <sub>2</sub> CH <sub>2</sub> OC <sub>2</sub> H <sub>5</sub>	(4-)F	61
53	1	(2-)S	H	-OCH 	(4-)F	

Table 1 - Continuation

5

Ex. No.	n	(Position) Q	R <sup>1</sup>	R <sup>2</sup>	(Position) X	Melting point (°C)
54	1	(2-)S	H	$  \begin{array}{c}  \text{CH}_3 \\    \\  -\text{CH}_2-\text{C}-\text{C}-\text{Cl} \\  \quad \quad   \\  \quad \quad \text{Cl}  \end{array}  $	(4-)F	113
55	1	(2-)S	H	$-\text{CH}_2\text{CH}_2\text{OCH}_3$	(4-)F	81
56	1	(2-)S	H	$  \begin{array}{c}  \text{CH}_3 \\  \diagup \\  -\text{CH} \\  \diagdown \\  \text{CF}_3  \end{array}  $	(4-)F	45
57	1	(2-)S	H	$  \begin{array}{c}  -\text{CHC}_3\text{H}_7 \\    \\  \text{CH}_3  \end{array}  $	(4-)F	62
58	1	(2-)S	H	$-\text{OCH}_2\text{CH}=\text{CH}_2$	(4-)F	48
59	1	(2-)S	H	H	(4-)F	
60	1	(2-)S	H	$-\text{C}(\text{CH}_3)_3$	(4-)F	
61	1	(2-)S	H	$-\text{CH}(\text{CH}_3)_2$	(4-)F	99
62	1	(2-)S	H	$-\text{CH}_2\text{CH}(\text{CH}_3)_2$	(4-)F	
63	1	(2-)O	CH <sub>3</sub>	CH <sub>3</sub>	(4-)F	120
64	1	(2-)S	H	$-\text{CO}-\text{CH}_3$	(4-)F	
65	1	(2-)S	H	$-\text{CO}-\text{CH}(\text{CH}_3)_2$	(4-)F	80
66	1	(2-)S	H	$-\text{CO}-\text{C}_2\text{H}_5$	(4-)F	96
67	1	(2-)O	CH <sub>3</sub>	OH	(4-)F	154
68	1	(2-)O	H	CH <sub>3</sub>	(4-)F	193
69	0	(3-)O	CH <sub>3</sub>	CH <sub>3</sub>	-	136

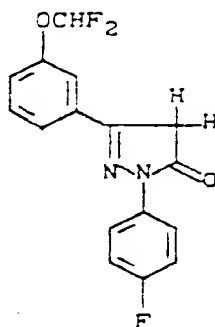
Table 1 - Continuation

5

Ex. No.	n	(Posi- tion) Q	R <sup>1</sup>	R <sup>2</sup>	(Posi- tion) X	Melting point (°C)
70	1	(3-)O	CH <sub>3</sub>	OH	(4-)F	146
71	1	(2-)S	CH <sub>3</sub>	OH	(2-)F	124
72	0	(3-)O	CH <sub>3</sub>	H	-	137
73	1	(2-)S	CH <sub>3</sub>	H	(2-)F	127
74	1	(3-)O	CH <sub>3</sub>	OH	(2-)F	78
75	1	(3-)O	CH <sub>3</sub>	H	(2-)F	126

Starting substances of the formula (II):

Example (II-1)

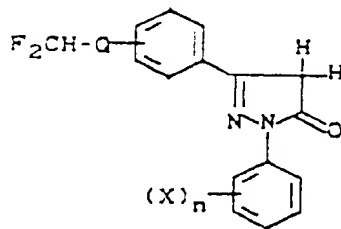


5 To a solution of 4.06 g (25 mmol) of 4-fluorophenyl-  
hydrazine hydrochloride in 100 ml of ethanol there are  
added 6.5 g (25 mmol) of ethyl (3-difluoromethoxy-  
benzoyl)-acetate and 2.05 g (25 mmol) of sodium acetate,  
and the reaction mixture is stirred for 20 hours at 20°C.  
10 It is then concentrated under a water pump vacuum, the  
residue is stirred with petroleum ether, and the crystal-  
line product is isolated by filtration with suction,  
washed with water and dried.

7.4 g (92% of theory) of 2-(4-fluorophenyl)-5-(3-di-  
fluoromethoxyphenyl)-2,4-dihydro-3H-pyrazol-3-one of  
15 melting point 114°C are obtained.

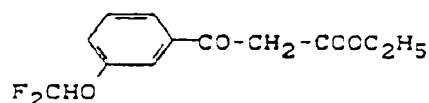
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Other compounds of the formula (II) which can be prepared analogously to Example (II-1) are those listed in Table 2 below.



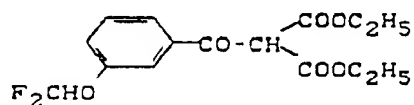
5 Table 2: Examples of the compounds of the formula (II)

Ex. No.	n	(Position) Q	(Position) X	Melting point (°C)
(II-2)	1	(2-)S	(4-)F	
(II-3)	1	(3-)S	(4-)F	78
(II-4)	1	(2-)O	(4-)F	
(II-5)	1	(2-)S	(4-)Cl	
(II-6)	2	(2-)S	(2,4-)F <sub>2</sub>	
(II-7)	1	(2-)S	(3-)Cl	
(II-8)	1	(3-)S	(3-)Cl	
(II-9)	1	(3-)S	(4-)CF <sub>3</sub>	
(II-10)	1	(2-)S	(2-)F	
(II-11)	1	(3-)O	(2-)F	
(II-12)	1	(3-)O	(4-)F	
(II-13)	1	(3-)O	H	

Starting substances of the formula (V):Example (V-1)

5 A mixture of 91.5 g (0.28 mol) of diethyl (3-difluoro-  
methoxybenzoyl)-malonate, 300 ml of water and 3 g of  
p-toluenesulphonic acid is refluxed for 8 hours. After  
cooling, the mixture is extracted with methylene chlor-  
ide, and the organic phase is stirred with saturated  
10 sodium hydrogen carbonate solution, dried with sodium  
sulphate and filtered. The filtrate is concentrated under  
a water pump vacuum, and the residue is distilled under  
an oil pump vacuum.

21.2 g (30% of theory) of ethyl (3-difluoromethoxy-  
benzoyl)-acetate of boiling point 118°C (at 0.01 torr)  
15 are obtained.

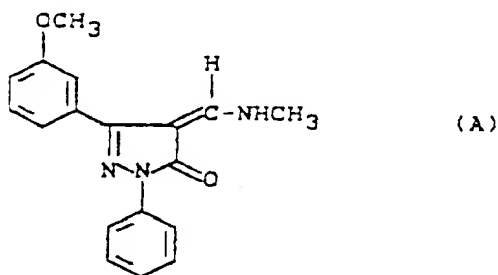
Starting substances of the formula (VII):Example (VII-1)

27.4 g (0.29 mol) of magnesium chloride are added in portions to 150 ml of acetonitrile, the internal temperature being kept below 25°C by external cooling with ice/water. After the mixture has cooled to -10°C, 46.1 g (0.29 mol) of diethyl malonate and 58.1 g (0.58 mol) of triethylamine are added dropwise in succession. After the mixture has been stirred at -10°C for a further 30 minutes, 59.5 g (0.29 mol) of 3-difluoromethoxy-benzoyl chloride, dissolved in 120 ml of acetonitrile, are added dropwise, and the mixture is stirred for 12 hours at 20°C. After 250 ml of 5N hydrochloric acid have been added, the mixture is extracted using methylene chloride, and the organic phase is dried using sodium sulphate and filtered. The solvent is carefully removed from the filtrate by distillation under a water pump vacuum.

92.4 g (97% of theory) of diethyl (3-(difluoromethoxy-benzoyl)-malonate as an oily residue of refractive index  $n_D^{24}$  : 1.4629 are obtained.

Use Examples:

In the Use Examples which follow, the compound shown below is used as comparison substance:



5-(3-Methoxyphenyl)-4-methylaminomethylene-2-phenyl-2,4-dihydro-3H-pyrazol-3-one  
(disclosed in EP-A 274,642).

5     Example A

Post-emergence test

Solvent:            5 parts by weight of acetone

Emulsifier:        1 part by weight of alkylaryl polyglycol  
ether

10     To produce a suitable preparation of active compound, 1 part by weight of active compound is mixed with the stated amount of solvent, the stated amount of emulsifier is added and the concentrate is diluted with water to the desired concentration.

15     Test plants which have a height of 5 - 15 cm are sprayed

with the preparation of the active compound in such a way as to apply the particular amounts of active compound desired per unit area. After three weeks, the degree of damage to the plants is rated in % damage in comparison to the development of the untreated control. The figures denote:

0% = no action (like untreated control)  
100% = total destruction

In this test, a powerful action against weeds is shown, for example, by the compounds of Preparation Examples 3, 5, 7, 8, 10, 11, 14, 15, 17, 19, 20, 26, 27, 32, 33, 36, 38, 40, 52, 55, 59 and 67, combined with good crop plant compatibility.

Example B

Pre-emergence test

Solvent: 5 parts by weight of acetone  
Emulsifier: 1 part by weight of alkylaryl polyglycol ether

To produce a suitable preparation of active compound, 1 part by weight of active compound is mixed with the stated amount of solvent, the stated amount of emulsifier is added and the concentrate is diluted with water to the

desired concentration.

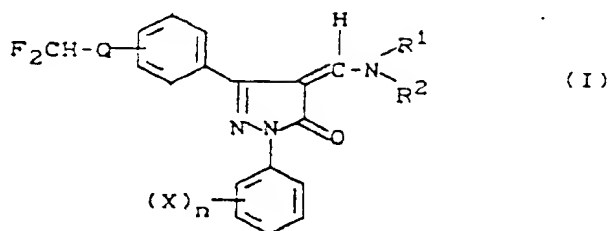
5       Seeds of the test plants are sown in normal soil and,  
after 24 hours, watered with the preparation of the  
active compound. It is expedient to keep constant the  
amount of water per unit area. The concentration of the  
active compound in the preparation is of no importance,  
only the amount of active compound applied per unit area  
being decisive. After three weeks, the degree of damage  
to the plants is rated in % damage in comparison to the  
10       development of the untreated control. The figures denote:

0% = no action (like untreated control)  
100% = total destruction

15       A clearly superior activity compared with the prior art  
is shown in this test, for example, by the compounds of  
the following Preparation Examples: 7 and 15.

What is claimed is:

1. A diarylpyrazolinone of the general formula (I)



wherein

- 5           n       represents the numbers 0, 1, 2 or 3,
- Q       represents oxygen or sulphur,
- R<sup>1</sup>     represents hydrogen or C<sub>1</sub>-C<sub>4</sub>-alkyl,
- R<sup>2</sup>     represents hydrogen, hydroxyl, amino, or a  
 10           radical from the series comprising C<sub>1</sub>-C<sub>5</sub>-alkyl,  
           C<sub>2</sub>-C<sub>5</sub>-alkenyl, C<sub>2</sub>-C<sub>5</sub>-alkinyl, C<sub>1</sub>-C<sub>5</sub>-hydroxyalkyl,  
           C<sub>1</sub>-C<sub>4</sub>-alkoxy-C<sub>1</sub>-C<sub>4</sub>-alkyl, C<sub>3</sub>-C<sub>6</sub>-cycloalkyl-C<sub>1</sub>-C<sub>4</sub>-  
           alkyl, phenyl-C<sub>1</sub>-C<sub>4</sub>-alkyl, C<sub>1</sub>-C<sub>6</sub>-alkoxy, C<sub>3</sub>-C<sub>6</sub>-  
           alkenyloxy, phenyl-C<sub>1</sub>-C<sub>2</sub>-alkoxy, C<sub>1</sub>-C<sub>6</sub>-alkyl-  
 15           amino, C<sub>1</sub>-C<sub>4</sub>-alkylcarbonylamino or di-(C<sub>1</sub>-C<sub>4</sub>-  
           alkyl)-amino, each of which is optionally  
           substituted by halogen, and

X represents hydrogen, halogen, or a radical from the series comprising C<sub>1</sub>-C<sub>4</sub>-alkyl or C<sub>1</sub>-C<sub>4</sub>-alkoxy, each of which is optionally substituted by halogen, with the exception of the compounds:

5 2-phenyl-, 2-(4-fluorophenyl)- and 2-(4-chlorophenyl)-5-(2-difluoromethylthiophenyl)-4-(N-hydroxy-methylaminomethylene)-2,4-dihydro-3H-pyrazol-3-one, 2-phenyl-, 2-(4-fluorophenyl)- and 2-(4-chlorophenyl)-5-(3-difluoromethylthiophenyl)-4-(N-hydroxy-methylaminomethylene)-  
10 2,4-dihydro-3H-pyrazol-3-one, 2-phenyl-, 2-(4-fluorophenyl)- and 2-(4-chlorophenyl)-5-(2-difluoromethylthiophenyl)-4-dimethylaminomethylene-2,4-dihydro-3H-pyrazol-3-one as well as 2-phenyl-, 2-(4-fluorophenyl)- and 2-(4-chlorophenyl)-5-(3-difluoromethylthiophenyl)-4-dimethylaminomethylene-2,4-dihydro-3H-pyrazol-  
15 3-one.

20 2. A diarylpyrazolinone of the general formula (I) according to Claim 1, wherein

n represents the numbers 0, 1 or 2,

Q represents oxygen or sulphur,

R<sup>1</sup> represents hydrogen, methyl or ethyl,

R<sup>2</sup> represents hydrogen, hydroxyl, amino, or

5

represents a radical from the series comprising  $C_1$ - $C_3$ -alkyl,  $C_3$ - $C_5$ -alkenyl,  $C_3$ - $C_5$ -alkinyl,  $C_1$ - $C_3$ -hydroxyalkyl,  $C_1$ - $C_2$ -alkoxy- $C_1$ - $C_2$ -alkyl,  $C_1$ - $C_4$ -alkoxy or  $C_3$ - $C_4$ -alkenyloxy, each of which is optionally substituted by fluorine and/or chlorine,

10

or represents a radical from the series comprising  $C_3$ - $C_5$ -cycloalkyl- $C_1$ - $C_2$ -alkyl, phenyl- $C_1$ - $C_2$ -alkyl, phenyl- $C_1$ - $C_2$ -alkoxy,  $C_1$ - $C_4$ -alkylamino,  $C_1$ - $C_4$ -alkylcarbonylamino or dimethylamino, each of which is optionally substituted by fluorine, chlorine and/or bromine, and

15

X represents hydrogen, fluorine, chlorine, bromine or a radical from the series comprising methyl, ethyl, methoxy or ethoxy, each of which is optionally substituted by fluorine and/or chlorine, with the exception of the compounds:

20

25

2-phenyl-, 2-(4-fluorophenyl)- and 2-(4-chlorophenyl)-5-(2-difluoromethylthiophenyl)-4-(N-hydroxy-methylaminomethylene)-2,4-dihydro-3H-pyrazol-3-one, 2-phenyl-, 2-(4-fluorophenyl)- and 2-(4-chlorophenyl)-5-(3-difluoromethylthiophenyl)-4-(N-hydroxy-methylaminomethylene)-2,4-dihydro-3H-pyrazol-3-one, 2-phenyl-, 2-(4-fluorophenyl)- and 2-(4-chlorophenyl)-5-(2-difluoromethylthiophenyl)-4-dimethylaminomethylene-2,4-dihydro-3H-pyrazol-3-one as well

as 2-phenyl-, 2-(4-fluorophenyl)- and 2-(4-chlorophenyl)-5-(3-difluoromethylthiophenyl)-4-dimethylaminomethylene-2,4-dihydro-3H-pyrazol-3-one.

5        3. A diarylpyrazolinone of the general formula (I) according to Claim 1, wherein  
n        represents the numbers 0, 1 or 2,

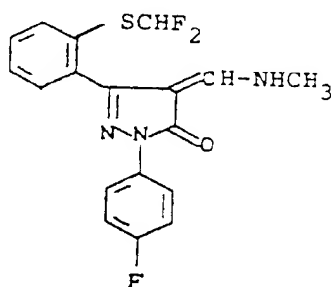
Q        represents oxygen or sulphur,

R<sup>1</sup>       represents hydrogen or methyl,

10       R<sup>2</sup>       represents hydrogen, hydroxyl, amino, or represents a radical from the series comprising methyl, ethyl, propyl, isopropyl, butyl, isobutyl, sec-butyl or tert-butyl, each of which is optionally substituted by fluorine, or  
15       represents allyl, propargyl, 1-methyl-propargyl, 1,1-dimethyl-propargyl, hydroxyethyl, hydroxypropyl, methoxyethyl, ethoxyethyl, cyclopropylmethyl, cyclohexylmethyl, cyclohexylethyl, phenylmethyl, phenylethyl, methoxy,  
20       ethoxy, propoxy, allyloxy, benzyloxy, methyl-amino, dimethylamino, acetylamino, propionyl-amino, butyrylamino or isobutyrylamino, and

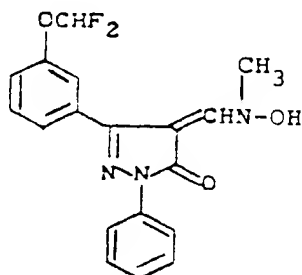
25       X       represents hydrogen, fluorine, chlorine or trifluoromethyl, with the exception of the compounds:

- 2-phenyl-, 2-(4-fluorophenyl)- and 2-(4-chlorophenyl)-5-(2-difluoromethylthiophenyl)-4-(N-hydroxy-methylaminomethylene)-2,4-dihydro-3H-pyrazol-3-one, 2-phenyl-, 2-(4-fluorophenyl)- and 2-(4-chlorophenyl)-5-(3-difluoromethylthiophenyl)-4-(N-hydroxy-methylaminomethylene)-2,4-dihydro-3H-pyrazol-3-one, 2-phenyl-, 2-(4-fluorophenyl)- and 2-(4-chlorophenyl)-5-(2-difluoromethylthiophenyl)-4-dimethylaminomethylene-2,4-dihydro-3H-pyrazol-3-one as well as 2-phenyl-, 2-(4-fluorophenyl)- and 2-(4-chlorophenyl)-5-(3-difluoromethylthiophenyl)-4-dimethylaminomethylene-2,4-dihydro-3H-pyrazol-3-one.
4. A compound according to claim 1, wherein such compound is 2-(4-fluorophenyl)-5-(2-difluoromethylthiophenyl)-4-methylaminomethylene-2,4-dihydro-3H-pyrazol-3-one of the formula



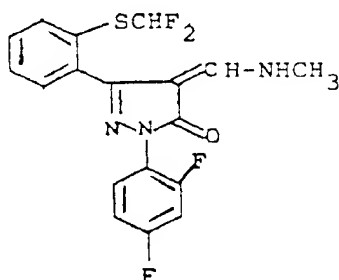
5. A compound according to claim 1, wherein such compound is

2-phenyl-5-(3-difluoromethoxyphenyl)-4-(N-methyl-N-hydroxyaminomethylene)-2,4-dihydro-3H-pyrazol-3-one of the formula



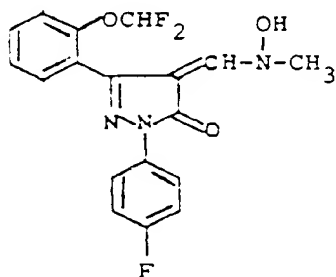
6. A compound according to claim 1, wherein such compound is

2-(2,4-difluorophenyl)-5-(2-difluorothiomethylphenyl)-4-methylaminomethylene-2,4-dihydro-3H-pyrazol-3-one of the formula



7. A compound according to claim 1, wherein such compound is

2-(4-fluorophenyl)-5-(2-difluoromethoxyphenyl)-4-(N-methyl-N-hydroxyaminomethylene)-2,4-dihydro-3H-pyrazol-3-one of the formula



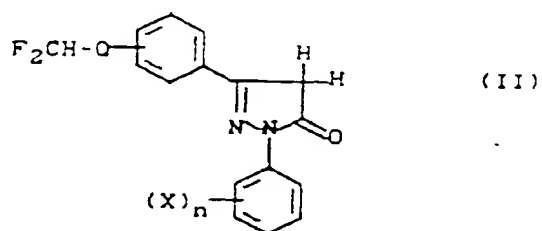
8. A herbicidal composition comprising a herbicidally effective amount of a compound according to claim 1 and a diluent.

9. A method of combating unwanted vegetation which comprises applying to such vegetation or to a locus from which it is desired to exclude such vegetation a herbicidally effective amount of a compound according to claim 1.

10. The method according to claim 9 wherein such compound is

2-(4-fluorophenyl)-5-(2-difluoromethylphenyl)-4-methylaminomethylene-2,4-dihydro-3H-pyrazol-3-one,  
 2-phenyl-5-(3-difluoromethoxyphenyl)-4-(N-methyl-N-hydroxyaminomethylene)-2,4-dihydro-3H-pyrazol-3-one,  
 2-(2,4-difluorophenyl)-5-(2-difluoromethylphenyl)-4-methylaminomethylene-2,4-dihydro-3H-pyrazol-3-one,  
 2-(4-fluorophenyl)-5-(2-difluoromethoxyphenyl)-4-(N-methyl-N-hydroxyaminomethylene)-2,4-dihydro-3H-pyrazol-3-one.

11. A pyrazolinone of the general formula (II)



wherein

n represents the numbers 0, 1, 2 or 3,

Q represents oxygen or sulphur and

5 X represents hydrogen, halogen, or a radical from the series comprising C<sub>1</sub>-C<sub>4</sub>-alkyl or C<sub>1</sub>-C<sub>4</sub>-alkoxy, each of which is optionally substituted by halogen.

12. An aroylacetate of the general formula (V)

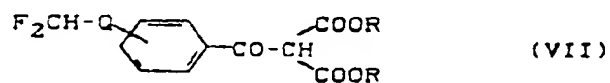


wherein

Q represents oxygen or sulphur and

R represents C<sub>1</sub>-C<sub>6</sub>-alkyl.

13. An arcylnalonic diester of the general formula (VII)



5

wherein

Q represents oxygen or sulphur and

R represents C<sub>1</sub>-C<sub>6</sub>-alkyl.

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